TECHNICAL REPORT ON YEAR ONE OF A TWO-YEAR SCREENING SURVEY OF CONTAMINANTS IN FISH FROM CALIFORNIA LAKES AND RESERVOIRS

Prepared for the Surface Water Ambient Monitoring Program

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EXECUTIVE SUMMARY

Objectives of the Study and This Report

This technical report presents results from the first year of a two-year screening survey of the potential for human exposure and health risks from consuming contaminated sport fish from California lakes and reservoirs. The survey is being performed as part of the State Water Resources Control Board's Surface Water Ambient Monitoring Program (SWAMP). This effort marks the beginning of a new long-term, statewide, comprehensive bioaccumulation monitoring program for California surface waters.

The Lakes Survey was designed to answer three questions:

- 1. What is the condition of California lakes with respect to contaminants in sport fish?
- 2. Should a specific lake be considered for inclusion on the 303(d) list due to bioaccumulation of contaminants in sport fish?
- 3. Should additional sampling of contaminants in sport fish at a lake be conducted for the purpose of developing consumption guidelines?

The results presented in this report provide a preliminary assessment of the statewide scope of the bioaccumulation problem in California lakes and reservoirs. The report also provides lake-specific information that can be used to establish priorities for cleanup actions, and identifies lakes where additional sampling may be needed to support fish consumption advisories. The report does not make specific recommendations for 303(d) listing, as these decisions are made by the State and Regional Water Boards. However, the database generated by this effort is tailored to providing the information that the Boards will need to make listing determinations.

This report is intended for a technical audience (agency staff, scientists, and peer reviewers). A summary for a non-technical audience will be prepared separately. In 2010, a final report on the Lakes Survey will be prepared that will cover both years of sampling and a more detailed exploration of factors influencing patterns in bioaccumulation, including sources of contamination.

Sampling Design

The overall goal of this screening study is to determine whether or not fish in California lakes have concentrations of contaminants that exceed thresholds for protection of human health. Fish tissue samples were collected from both targeted and randomly selected lakes throughout the state. The study focused on sampling indicator species that tend to accumulate high concentrations of the contaminants of concern. Black bass (including largemouth, smallmouth, and spotted bass) and Sacramento pikeminnow were the key indicator species for methylmercury. Channel catfish and common carp were the primary indicators for organic

pollutants. In the first year of this screening study, over 6000 fish from 18 species were collected from 152 lakes and reservoirs in California.

Overall Condition Assessment

Sport fish tissue concentrations were evaluated using thresholds developed by the California Office of Environmental Health Hazards Assessment (OEHHA) for methylmercury, PCBs, dieldrin, DDTs, chlordanes, and selenium. Lakes were considered "clean" if all average pollutant concentrations in all species were below all OEHHA thresholds. Only 15% of the lakes sampled in 2007 were in the "clean" category.

Furthermore, whether these lakes are entirely clean depends upon whether highmethylmercury species such as largemouth bass or self-sustaining trout populations are really absent from these lakes. Nevertheless, falling into the green category in this survey is a positive outcome indicating that the most readily caught species in a lake have pollutant concentrations that are below thresholds for concern. These lakes can be considered to be low priorities for monitoring to support development of fish consumption advisories. Methylmercury was the pollutant primarily responsible for the remaining 85% of lakes having at least one species with an average concentration above thresholds.

Methylmercury

Methylmercury is the pollutant that poses the most widespread potential health risks to consumers of fish caught from California lakes. Overall, 74% of the 152 lakes sampled had a fish species with an average methylmercury concentration above the threshold at which OEHHA would consider recommending consumption of less than three servings per week (0.07 ppm). Approximately 26% of the 152 lakes surveyed had a species with an average concentration high enough that OEHHA would consider recommending <u>no</u> consumption of the contaminated species (greater than 0.44 ppm). These lakes should be considered high priorities for further monitoring in support of consumption advisory development and management actions.

Methylmercury concentrations across the state varied at a regional scale. In northern California, low concentrations were commonly observed in high elevation (above 2000 ft) lakes in the Sierra Nevada and Trinity Alps. The highest species averages observed in these lakes were usually below 0.07 ppm. Trout were the most commonly caught species in these lakes, and tend to exhibit lower methylmercury concentrations than largemouth bass. In contrast, methylmercury concentrations in largemouth bass and other species in lower elevation (below 2000 ft) lakes in northern California were almost always higher than 0.07 ppm, and half of these lakes were higher than 0.44 ppm.

Although methylmercury concentrations were generally not as high in southern California, the methylmercury problem is not confined to northern

California and its well-known mining regions. Most of the 55 lakes in southern California (69%) were above 0.07 ppm. The majority were between 0.07 and 0.44 ppm (55%), but 15% had a species average above 0.44 ppm.

PCBs

PCBs were second to methylmercury in reaching concentrations posing potential health risks to consumers of fish caught from California lakes. Approximately 37% of the lakes had a fish species with an average PCB concentration above the lowest OEHHA threshold (3.6 ppb). In contrast to methylmercury, only 1% of the lakes sampled had a species with an average concentration high enough that OEHHA would consider recommending no consumption of the contaminated species (120 ppb).

Southern California was the region with the highest PCB concentrations, with 60% of lakes above 3.6 ppb. In northern California, low concentrations were commonly observed in high elevation lakes in the Sierra Nevada and Trinity Alps (only 7% of lakes were above 3.6 ppb), and concentrations were generally greater in lower elevation lakes (41% of lakes above 3.6 ppb).

Other Pollutants

Concentrations of dieldrin, DDT, chlordane, and selenium were generally low, and infrequently exceeded OEHHA thresholds. The high elevation lakes of northern California never exceeded any OEHHA threshold for these pollutants.

Risks to Wildlife

The OEHHA thresholds do not take into consideration effects of contaminants on wildlife. Risks to wildlife, such as fish-eating birds, at the concentrations observed in California lakes, are likely to be higher than for humans in some instances. Assessment of the impact of bioaccumulation on aquatic life, though not feasible with the current level of funding for this program, is considered a significant concern and would be evaluated if funding increases sufficiently in the future.

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INTRODUCTION

This document presents results from the first year of a two-year screening survey of bioaccumulation in California lakes and reservoirs. This work is being performed as part of the State Water Resources Control Board's Surface Water Ambient Monitoring Program (SWAMP). This effort will mark the beginning of a new long-term, statewide, comprehensive bioaccumulation monitoring program for California surface waters.

Oversight for this project is being provided by the SWAMP Roundtable. The Roundtable is composed of State and Regional Board staff and representatives from other agencies and organizations including USEPA, the Department of Fish and Game, the Office of Environmental Health Hazard Assessment (OEHHA), and the University of California. Interested parties, including members of other agencies, consultants, or other stakeholders also participate.

The Roundtable has formed a subcommittee, the Bioaccumulation Oversight Group (BOG) that focuses on SWAMP bioaccumulation monitoring. The BOG is composed of State and Regional Board staff and representatives from other agencies and organizations including USEPA, the Department of Fish and Game, the Office of Environmental Health Hazard Assessment, and the San Francisco Estuary Institute. The members of the BOG possess extensive experience with bioaccumulation monitoring.

The BOG has also convened a Bioaccumulation Peer Review Panel that is providing evaluation and review of the bioaccumulation program. The members of the Panel are internationally-recognized authorities on bioaccumulation monitoring.

The BOG has developed and begun implementing a plan to evaluate bioaccumulation impacts on the fishing beneficial use in all California water bodies. Sampling of sport fish in lakes and reservoirs has been conducted in the first two years (2007 and 2008). In 2009 and 2010, sport fish from the California coast, including bays and estuaries will be sampled. Sport fish from rivers and streams will be sampled in 2011. In 2012 the plan is to again begin a two year effort on lakes and begin another five-year cycle of sampling these water body types.

The Lakes Survey

Management Questions for this Survey

Three management questions were articulated to guide the design of the Lakes Survey. These management questions are specific to this initial monitoring effort; different sets of management questions will be established to guide later efforts.

Management Question 1

What is the condition of California lakes with respect to bioaccumulation in sport fish?

Answering this question has been the goal of assessments related to section 305(b) of the federal Clean Water Act (CWA). In the past, 305(b) reports have provided water quality information to the general public and served as the basis for U.S. EPA's National Water Quality Inventory Report to Congress. The report provided a statewide, comprehensive assessment of the status of California water bodies with respect to support of designated beneficial uses (e.g., SWRCB 2003). In the future, this information will be part of an "Integrated Report" formally known as the California CWA Section 305(b)/303(d) Integrated Report. This report will satisfy both the CWA section 305(b) and section 303(d) requirements (CWA section 303(d) is discussed further below). Answering this question also provides the state and the public with information that helps describe the magnitude, spatial dimensions, and priority of the bioaccumulation problem relative to other environmental and societal problems.

The information needed to answer this question is the representative, average concentration of bioaccumulative contaminants in each lake for an adequately large sampling of lakes.

Management Question 2

Should a specific lake be considered for inclusion on the 303(d) list due to bioaccumulation of contaminants in sport fish?

Answering this question is critical to determining the need for 303(d) listing and cleanup actions to reduce contaminant exposure in specific water bodies. Total Maximum Daily Load evaluations (TMDLs) are required for water bodies placed on the 303(d) list. This is the principal regulatory mechanism being used by the State Water Board, the Regional Water Boards, and USEPA to establish priorities for management actions.

The State Board has established a Listing Policy for placing water bodies on the CWA Section 303(d) list. The Listing Policy establishes a standardized approach and includes California listing and delisting factors. The fish tissue information needed to make a listing determination depends on the type of data and the pollutant. The more representative the samples are of the water body, the better. The goal in addressing Management Question 2 in this survey was to assist the Regional Boards and State Board by providing the data needed for listing decisions. Section 303(d) listing decisions will be made by the Regional Boards using the data generated in the Lakes Survey.

Management Question 3

Should additional sampling of bioaccumulation in sport fish at a lake be conducted for the purpose of developing consumption guidelines?

Answering this question is essential as a first step in determining the need for more thorough sampling in support of developing consumption guidelines. Consumption guidelines provide a mechanism for reducing human exposure in the

near-term. The information requirements for consumption guidelines are more extensive than for 303(d) listing. OEHHA, the agency responsible for issuing consumption guidelines, needs samples representing 9 or more fish from a variety of species abundant in a water body in order to issue guidance. It is useful to have information not only on the species with high concentrations, but also the species with low concentrations so anglers can be encouraged to target the low species.

Overall Approach

The overall approach taken to answer these three questions was to perform a statewide screening study of bioaccumulation in sport fish. The highest priority for SWAMP in the short-term is to answer Management Questions 1 and 2. Answering these questions will provide a basis for decision-makers to understand the scope of the bioaccumulation problem and will provide regulators with information needed to establish priorities for cleanup actions. As a next step, developing consumption guidelines that inform the public on ways to reduce their exposure is also a high priority, and this initial monitoring effort is cost-effectively establishing a foundation for this by identifying lakes that are candidates for additional sampling in support of guideline development.

It is anticipated that the screening study will lead to more detailed followup investigations of many water bodies that become placed on the 303(d) list or where consumption guidelines are needed.

This Report

The purpose of this technical report, which presents results from the first year of the Lakes Survey, is to provide agency staff, scientists, and peer reviewers with a summary of initial findings and a basis for technical evaluation of the work. A nontechnical summary of this work for a general audience will be prepared separately. Since this report only covers a partial dataset, a limited amount of interpretation of the patterns observed has been performed. In 2010, a final report on the lakes survey will be prepared that will cover both years of sampling and a more detailed exploration of factors influencing patterns in bioaccumulation, including sources.

METHODS

Sampling Design

The sampling plan was developed to address the three management questions for the project. In 2007, sampling was conducted at 152 lakes and reservoirs across the state (Figures 1a-c, Tables 1a,b). Targeted sampling of "popular" lakes comprised the bulk of the year 1 effort (102 of 152), with the remainder comprising a random sampling. A list of the 216 most popular fishing lakes and reservoirs in California was compiled, as identified through a review of published fishing guides (Stienstra 2004), websites, and consultation with Regional Board staff. In 2007, 80 of these lakes were sampled in

random order, using the generalized random tessellation-stratified (GRTS) approach developed for USEPA's Environmental Monitoring and Assessment Program (Stevens and Olsen 2004). The remaining popular lakes were sampled in 2008 (the 2008 samples are currently being analyzed). In the random selection of these lakes, each lake was assigned an equal probability of inclusion. The advantage of this approach is that if the entire population of 216 lakes is not sampled, inferences can still be drawn about the population as a whole, including the unsampled popular lakes.

In addition to the statewide targeted sampling of popular lakes, this report also includes data obtained from a coordinated targeted sampling of lakes in Region 4 (Figures 1a,c). Region 4 augmented the statewide effort with funds to provide for sampling of 22 additional lakes, including a more thorough analysis of replicate samples than was feasible in the statewide effort.

The second major emphasis of sampling in 2007 was to provide an evaluation of statewide lake condition. A randomized sampling of 50 lakes from the entire population of California lakes was conducted to provide an unbiased statewide assessment, and a valuable frame of reference for interpreting bias in the targeted sampling. However, many of the lakes and reservoirs in California are inaccessible or unfishable. To avoid wasting sampling resources on these lakes, the population of random lakes was restricted to lakes greater than 4 ha in size that could be accessed and sampled within a one day period. Furthermore, given the general focus of the survey on evaluating the impact of bioaccumulation on the fishing beneficial use, higher inclusion probabilities were assigned to larger lakes. These restrictions resulted in the exclusion of many lakes from the sample population. As with the popular lakes, the 50 random lakes were selected using the GRTS approach. The Sampling Plan (Davis et al. 2007a) provides more details on the design.

Target Species

The overall goal of this screening study is to determine whether or not California lakes have concentrations of contaminants that are above thresholds for protection of human health. Therefore, the study focused sampling on indicator species that tend to accumulate the highest concentrations of the contaminants of concern. Primary target species were selected that are popular for human consumption (e.g., rainbow trout [Oncorhynchus mykiss]), and/or are effective at documenting spatial trends in methylmercury (e.g., largemouth bass [Micropterus salmoides]) or organics (e.g., common carp [Cyprinus carpio]). Methylmercury biomagnifies primarily through its accumulation in muscle tissue, so top predators such as largemouth bass tend to have the highest methylmercury concentrations. In contrast, organic contaminants are biomagnified through accumulation in lipid. Bottom-feeding species such as channel catfish (Ictalurus punctatus) and common carp tend to have the highest lipid concentrations in their muscle tissue, and therefore usually have the highest concentrations of organics. Consequently, this study targeted two indicator species in each lake – a top predator (e.g., black bass) as a methylmercury indicator and a high lipid, bottom feeding species (e.g., channel catfish or common carp) as an organics and

selenium indicator. Another advantage of this approach is that it provides a characterization of both the pelagic and benthic food chains. Notably, some high elevation lakes only had one abundant high trophic level species (i.e., a trout species). In these cases, the one species still represented a worst-case indicator and was sampled and analyzed for all of the pollutants on the analyte list. The species sampled most frequently were the primary target species: largemouth bass, common carp, and rainbow trout (Table 2). Other species were collected where the primary targets could not be obtained.

Specific size ranges for each species were established (Davis et al. 2007a). Sizes collected for each species are listed in Table 2. Black bass (including largemouth, smallmouth [Micropterus dolomieui], and spotted bass [Micropterus punctulatus]) and Sacramento pikeminnow (Ptychocheilus grandis) were the key methylmercury indicators. These species have a high trophic position and a strong size:methylmercury relationship. For these species, fish were sampled across a wide range of lengths and analyzed as individuals, to facilitate an ANCOVA of size-standardized methylmercury concentrations (however ANCOVA results are only presented for largemouth bass in this report). Individuals were analyzed for methylmercury in a few other instances for common carp (1 fish), kokanee (Oncorhynchus nerka, 1 fish), and striped bass (Morone saxatilis - 3 fish). As mentioned above, in many high elevation lakes only trout species were available. Furthermore, past sampling of rainbow trout in the Bay-Delta watershed found low concentrations and a weak size:methylmercury relationship in hatchery fish (Grenier et al. 2007, Melwani et al. 2007). Therefore, ANCOVA was not used for the trout species sampled in this survey (including rainbow, brown [Salmo trutta], and Eagle Lake trout [Oncorhynchus mykiss aguilarum]). Methylmercury was analyzed in composites of 5 individuals. These trout composites were also analyzed for organic contaminants. The size ranges established for trout were based on a combination of sizes prevalent in past sampling (Melwani et al. 2007) and the 75% rule recommended by USEPA (2000) for composite samples.

Channel catfish and common carp were the primary targets for high lipid bottom-feeders. These species were analyzed for organics, selenium, and methylmercury. Organics were expected to be highest in these species based on past monitoring in the Toxic Substances Monitoring Program and other studies (Davis et al. 2007b). Selenium was expected to be highest in these species, although the difference was not expected to be as distinct as for the organics, based on data from the Grassland Bypass Project (SFEI 2008). Methylmercury was expected to be highest in the pelagic predators, but concentrations are also expected to be above thresholds for concern in the bottom-feeders, so methylmercury was analyzed in these samples as well. Samples for these species were analyzed as composites (Table 2). The size ranges established for bottom-feeders were based on a combination of sizes prevalent in past sampling (Melwani et al. 2007) and the 75% rule recommended by USEPA (2000) for composite samples. In some lakes only bass were collected. In these cases, composites of the bass samples were created for organics analysis following the same approach (specified size range and the 75% rule) used for the bottom-feeders.

Locations Targeted

Lakes and reservoirs in California vary tremendously in size, from hundreds of small ponds less than 10 ha to Lake Tahoe at 50,000 ha. For larger lakes it is necessary to sample more than one location to obtain a representative characterization of the water body. In addition, it was frequently necessary to sample over a linear course of 0.5 - 1 mile to obtain the desired number of fish. Therefore, sampling locations in this study can be thought of as a circle with a diameter of 1 mile. For small lakes less than 500 ha in size, one sampling location covered a significant fraction of the surface area of the lake. However, for larger lakes, sampling of additional locations was performed. For lakes of medium size (500 - 1000 ha), two locations were generally sampled. For lakes in the large category (1000 - 5000 ha) and extra large category (>5000 ha), two to four locations were sampled.

Archiving Strategy

Due to the large number of water bodies to be sampled and an expectation that some of these would be below thresholds of concern, an archiving strategy was developed for composite samples of the bottom-feeder species. Individual samples of the predator species were analyzed for methylmercury only and an archiving strategy was not used. This decision was driven by the low cost of methylmercury analysis and the need for the largest dataset possible for statistical techniques, as described below. The archiving strategy for composite samples varied somewhat by the size of lake. For small lakes, two composites were collected to represent the entire lake area. Both composites were analyzed immediately for methylmercury, given the low cost of analysis. However, the second composite sample was only analyzed for organics and/or selenium if the first composite sample exceeded a threshold. The threshold for this follow-up analysis was designated as 75% of the threshold for concern (Table 3). These thresholds were based on a draft report by OEHHA. [NOTE: In OEHHA's final report (Klasing and Brodberg 2008) the thresholds were modified (Table 4). These newer thresholds were used for assessing the data in this report.] For lakes of larger size, composite samples were collected from each discrete location (the number of locations was based on lake size as described above). These composites were homogenized and analyzed immediately for methylmercury, but archived for organics and selenium. Aliquots of homogenate from each location composite were pooled to form a lake-wide composite. The lake-wide composite was analyzed immediately for organics and selenium. If the lake-wide composite concentration of any of the organics or selenium exceeded the threshold for follow-up analysis, then all of the discrete location composites were analyzed. This approach avoided expenditure of funds on organics analysis where it was not needed. Aliquots from all composites were archived whether they were analyzed or not, in case of any analytical problems or other circumstances calling for analysis or re-analysis at a later time

Field Sampling

Sport fish were collected from lakes across the state from June through November 2007 (Figures 1a-c, Tables 1a,b). Fish were collected by Moss Landing Marine

Laboratories (MLML) and the California Department of Fish and Games Water Pollution Control Laboratory (WPCL) staff with electrofisher boats and gill nets. The crew remained on location until the desired number of target species was caught. Total length (longest length from tip of tail fin to tip of nose/mouth), fork length (longest length from fork to tip of nose/mouth), and weight were measured in the field when possible; otherwise these parameters were measured in the lab and this was noted in the database. Latitude and longitude were recorded for every fish collected to document the spatial resolution among locations within a lake. Fish samples were wrapped in aluminum foil and frozen on dry ice for transportation to the laboratory. A Google Earth map of the sampling locations is available from the authors (contact Jay Davis, jay@sfei.org).

Sample Processing

Fish were stored at -20°C in their original bags until dissection and homogenization. Homogenates were also frozen until analysis was performed. Dissection and compositing of muscle tissue samples were performed following USEPA guidance (USEPA 2000). At the time of dissection, fish were placed in a clean lab in their original bags to thaw. After thawing, fish were cleaned by rinsing with de-ionized (DI) and ASTM Type II water, and were handled only by personnel wearing polyethylene or powder-free latex gloves (glove type is analyte dependent). All dissection materials were cleaned by scrubbing with Micro® detergent, rinsing with tap water, DI water, and finally ASTM Type II water. All fish were dissected skin-off, and only the fillet muscle tissue was used for analysis.

The labs analyzed the predator species as individuals for methylmercury and composites for organics, and trout and bottom species as composites. For composite samples, a subsample of equal mass was taken from each of 5 individual fish following the 75% size rule recommended by USEPA (2000). Tissue was homogenized with a Büchi B-400 mixer, to form a location composite with a target weight of 200g or greater. A subsequent lake-wide composite was created from equal portions of each contributing location composite within each lake. Post-homogenization aliquots were taken from the lake-wide composite for methylmercury, selenium, and organics analyses. Aliquots for methylmercury and selenium were transferred to pre-cleaned 30ml polypropylene jars. Organics aliquots were transferred to 60 ml borosilicate cleaned jars.

Scales were taken from all black bass individuals and analyzed for age by the counting of growth rings according to the methods found in Campana (2001). These results are in the database generated for this Survey, but not reported in this report. To obtain these data please contact Jay Davis (jay@sfei.org).

Archiving

Aliquots of homogenates of all composite samples analyzed were archived on a short-term basis to provide for reanalysis in case of any mishaps or confirmation. In addition, aliquots of the lakewide homogenates prepared for the bottom-feeder species were made and archived for long-term storage. This will provide a integrative,

representative sample for each lake that can be reanalyzed in later years to confirm earlier analyses, look for new chemicals of concern, provide material for application of new analytical methods, provide material for other ecological research, and other purposes. Long-term archiving of the lakewide homogenates is the most cost-effective approach to addressing this need.

Black bass individuals were archived on a short-term basis wrapped in the original aluminum foil. Long term archives, stored un-homogenized in glass, were created for the 5 individuals within the 75% size rule. The exception to this was when bass composites were created from the lake for organic analysis (when bottom-feeder species were not collected).

Furthermore, long-term archives were created for individuals of all species collected at those lakes identified for potential future trend analysis (Table 3). Each region identified lakes they were interested in sampling more often and establishing a baseline for trend analysis. A list of Trend Lakes can be found in Table 3 of the Lakes Survey Workplan (Davis et al. 2007). Collections and analyses did not differ at these lakes than at lakes not identified for trend analysis, however the archiving strategy was more intense. Trend lakes have individual archives retained for all species and all locations, and where sufficient tissue was present, location and lakewide archives were also retained. Otoliths were extracted from all individuals collected from each of the Trend Lakes. Otoliths were preserved in alcohol and stored in cryo-vials for preparation and reading at a later date if funds become available.

Chemical Analysis

Methylmercury and Selenium

Nearly all (>95%) of the mercury present in fish is methylmercury (Wiener et al. 2007). Consequently, monitoring programs usually analyze total mercury as a proxy for methylmercury, as was done in this study. USEPA (2000) recommends this approach, and the conservative assumption be made that all mercury is present as methylmercury to be most protective of human health.

Total mercury and selenium in muscle tissue were measured by Moss Landing Marine Laboratory (Moss Landing, CA).

All samples, blanks, and standards were prepared using clean techniques. ASTM Type II water and analytical grade chemicals were used for all standard preparations. A continuing calibration verification (CCV) was performed after every 10 samples. Samples whose initial or continuing calibration verification values drifted by more than $\pm 20\%$ of the true value were reanalyzed. One to three blanks (depending on analyte), a certified reference material (DORM-2), as well as a method duplicate and matrix spike pairs were run with each analytical batch of samples.

Total mercury in composite samples and individuals were analyzed by Thermal Decomposition, Catalytic Conversion, Amalgamation and Atomic Absorption Spectrophotometry which is described in EPA 7473 (USEPA, 1998) using a Direct Mercury Analyzer (Milestone DMA-80). Approximately 0.1-0.2 g of tissue was removed from either the composite homogenate or individual fillet, weighed and placed into the DMA-80 sample boat. Each sample is ultimately decomposed at 1000°C and the mercury is detected by a single beam spectrophotometer with sequential flow through two measurement cells. Samples were divided into analytical batches of 20 samples plus analytical QA samples (CRM, matrix spike and spike duplicate, duplicate and method blanks). Detection limits for total mercury and all of the other analytes are presented in Table 5.

Approximately 1.25 g of tissue from each composite sample for selenium analysis was weighed and digested by Microwave Assisted Acid Digestion (EPA 3052m) with concentrated nitric acid under pressure at 195°C. Samples were divided into analytical batches of 20 samples plus analytical QA samples (CRM, matrix spike and spike duplicate, duplicate and method blanks) digested simultaneously. Digestates were subsequently analyzed according to EPA 200.8 (USEPA, 1994) by Inductively Coupled Plasma-Mass Spectrometry (Perkin-Elmer ELAN 9000 ICP-MS).

Organics

Trace organics in muscle tissue were measured by the California Department of Fish and Game Water Pollution Control Laboratory (Rancho Cordova, CA).

Pressurized fluid extraction (EPA 3545A) was used for the extraction of organochlorine (OCs) pesticides and polychlorinated biphenyls (PCBs) in fish tissue. Gel permeation chromatography (EPA 3640A) and Florisil column chromatography (EPA 3620C) were used to purify and fractionate the extracts prior to analysis. Gas chromatography with triple quadrupole mass spectrometry (GC-MSMS) was used to analyze OC pesticides and PCBs. Dual column gas chromatography with dual electron capture detectors (GC-ECD) is used to analyze a small list of the more polar target OC pesticides.

Tissue samples containing surrogate compounds were extracted twice using a Dionex Accelerated Solvent Extractor (ASE 200) extractor. A portion of the extract was removed for percent lipid determination. Initial sample cleanup was done by gel permeation (size exclusion) chromatography. Additional cleanup and fractionation were done using Florisil® column chromatography.

A Varian Model 3800/1200L gas chromatograph (GC)/triple quadrupole mass spectrometer equipped with a Model 1177 split-splitless injector with electronic pressure control (EPC) and CombiPal® autosampler was used for all GC-MSMS analyses. The GC is equipped with a J&W Scientific 60 meter, 0.25 mm ID, 0.25 μm (film thickness) XLB column. The injector is operated isothermal at 280 degrees C in splitless mode with

pressure pulse (45 psi for 1.05 min). The mass spectrometer is operated in electron impact (EI) ionization MSMS mode using argon as the CID gas. Precursor and product ions were selected to optimize selectivity and sensitivity. Internal standard calibration using carbon 13 isotope labeled pesticides and PCB congeners were used.

An Agilent 6890plus gas chromatograph equipped with two 63Ni micro-electron capture detectors with EPC and autosampler was used to analyze a select list of the more polar pesticides. Two 60 meter, 0.25 mm ID, 0.25 μ m (film thickness) fused silica columns (J&W) were used. The injector is operated in splitless mode isothermal at 240 degrees C. Helium is used as the carrier gas at a linear velocity of 35 cm/sec. Nitrogen is used for the detector makeup at 30 mL/min.

Each analysis sequence included a minimum of seven calibration standards. The calibration curve concentration for chlorinated hydrocarbons was 0.5 ppb to 500 ppb. The calibration curve concentration range for polychlorinated biphenyl congeners (PCBs) was 0.5 ppb to 100 ppb. Higher concentrations of PCB standards (50 ppb to 1000 ppb) were analyzed with samples containing higher concentrations of PCBs.

An initial calibration blank and initial calibration verification standard were analyzed after the calibration standards and prior to the first sample extract. Continuing calibration blanks (CCBs) and calibration verification standards (CCVs) were analyzed after ten sample extracts. The CCV analyte concentrations were at the mid-range of the calibration curve (5-10 ppb).

A procedural blank, blank spike, matrix spike, matrix spike duplicate, sample duplicate and standard reference material (SRM 1588b-cod liver oil) produced and distributed by the National Institute of Standards and Technology (NIST) was extracted and analyzed with each set of 18 samples. Results of the QC analyses (except the ICVs and CCVs) are evaluated and reported with the data.

PCBs are reported as the sum of 55 congeners (Table 5). Concentrations in many lakes were near or below limits of detection (Table 5). The most abundant congeners were detected in 75 – 85% of the 252 samples analyzed for PCBs. Reporting frequencies were lower for some of these congeners due to blank contamination and other QA issues (note that Table 5 summarizes frequencies for the entire 152 lake dataset, including 14 samples that are being reanalyzed due to censoring of too many congeners to obtain an accurate sum of PCBs). Frequencies of detection and reporting were lower for the less abundant PCB congeners. The relative abundances of the PCB congeners fell within expected ranges, with some samples showing greater influences of Aroclor 1248 (San Luis Reservoir, Silverwood Lake, O'Neill Forebay, Lake Elsinore, Castaic Lake), Aroclor 1254 (Pyramid Lake, Peck Road Water Conservation Park, Alondra Park Lake), Aroclor 1260 (Chesbro Reservoir, Thermalito Afterbay, Hollenbeck Park Lake, Lake Chabot-San Leandro, Yosemite Lake), and Aroclor 1262 (Lake Chabot-Vallejo, Santa Fe Reservoir).

As recommended by USEPA (2000), DDTs are reported as the sum of six isomers and metabolites: p,p'-DDE, o,p'-DDE, p,p'-DDD, o,p'-DDD, p,p'-DDT, and o,p'-DDT. p,p'-DDE, the most abundant DDT isomer, was detected and reported in 95% of the 248 samples analyzed (Table 5). p,p'-DDD was detected second most frequently (76%). The other isomers and metabolites were detected in less than half of the samples. None of the DDT results were censored due to QA issues. The relative concentrations of the DDTs fell within expected ranges. The largest contribution of p,p'-DDT to the sum of DDTs was 17% at Lake Piru.

As recommended by USEPA (2000), chlordanes are reported as the sum of five components of technical chlordane: *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor, and oxychlordane. Concentrations in many lakes were near or below limits of detection (Table 5). The most abundant chlordane (*trans*-nonachlor) was detected in 79% of the 248 samples analyzed for chlordanes. Reporting frequencies were lower for some of the chlordanes due to blank contamination and other QA issues (note that Table 5 summarizes frequencies for the entire 152 lake dataset, including 4 samples that are being reanalyzed due to censoring of too many congeners to obtain an accurate sum of chlordanes). The relative abundances of the chlordanes fell within expected ranges.

In calculating sums of PCBs, DDTs, and chlordanes, results below detection limits were set to zero.

Quality Assurance

The 2007 samples were digested and analyzed in multiple batches. Batches consisted of 20 samples per batch. QAQC samples for the SWAMP Data Quality Objectives (DQOs) (precision, accuracy, recovery, completeness, and sensitivity) are performed for each batch as required by the SWAMP BOG QAPP (Bonnema, 2007). DQOs are reviewed and appropriate batch qualifiers assigned by the SWAMP Data Management Team. Measurement Quality Objectives were assessed according to the SWAMP BOG QAPP (see Table 12a and 12b in Bonnema [2007]).

A brief summary of the QA results is provided below. A more detailed summary is presented in Appendix 4. Data were classified as compliant, estimated, and rejected. Rejected data were not included in this report; compliant and estimated data were included and uploaded to the SWAMP Tissue Database 2.5.

A total of 22 samples did not pass QA review for all pollutants and were rejected. Data for lakes with rejected samples are not presented in this report. These samples are being reanalyzed and the data for these lakes will be reported in the final report for the Lakes Survey. Blank contamination issues for PCBs and chlordanes caused these rejections. These results were rejected when the affected samples had a summed value (either sum of PCBs or sum of chlordanes) higher than the FCG and where the final sum was reduced by 30% due to rejection of individual analytes (e.g., PCB congeners).

Blank Contamination

Blank matrices are run with each analytical batch to measure potential contamination of field samples from collection and sample handling. Acceptable blank results are those with values less than the method detection limit (MDL) for a particular analyte. Thirty-three analytes had some quantitative detection in the method blanks (4 pesticides, 4 PBDEs, 25 PCBs). Analyte concentrations in the field samples were compared to the associated method blank concentrations. Results for field samples that were less than 3 times the measured blank contamination were classified as rejected. The number of rejections in the dataset due to blank contamination was 1063 (including field samples, laboratory duplicates, and blind duplicates) while all other results were classified as compliant. Congeners that make up a significant percentage of the sum of PCBs or sum of chlordanes (PCB 101, PCB 110, PCB 118, PCB 138, PCB 149, PCB 153, PCB 180, *cis*-chlordane, *trans*-chlordane, *trans*-nonachlor) had rejections for some samples.

Accuracy

Certified Reference Materials (CRM), Matrix Spike/ Matrix Spike Duplicates (MS/D), and Laboratory Control Standards (LCS) are the QC elements used to assess the accuracy of an analytical method. Following SWAMP Management Quality Objectives, one QC accuracy element is allowed to fail in a batch and still be compliant. When more than one QC element fails, the analyte, for all batches, was classified as estimated. When the % Recovery was above 200 for more than 1 QC element, the analyte was rejected. In the case where there is only one QC element reported in the batch and the % Recovery was above 200 then the analyte would also be rejected. Two out of 165 total batches did not include MS/MSD performed at the required frequency (1 per batch of 20 samples). These two batches were classified as estimated. All 165 batches had the appropriate number of CRM and LCS per batch. Fifteen analytes had some accuracy failures (10 pesticides, 5 PCBs). No analytes were rejected due to accuracy measures.

Precision

Matrix Spike (MS)/Duplicates (MSD) and Laboratory duplicates (DUPs) were analyzed to assess laboratory precision. As required by the SWAMP BOG QAPP a duplicate of at least one field sample per batch was processed and analyzed. Three out of 165 total batches did not include DUPs performed at the required frequency and were classified as estimated. The duplicate results reported above the RL were compared and the Relative Percent Difference (RPD) was calculated. RPDs, for either the MS/MSD or DUPs, <25% were considered acceptable as specified in the QAPP. RPDs >25% but <50% were classified as estimated. RPDs >50% were classified as rejected. Rejections were applied to the entire batch for an analyte that failed precision. Thirty-four analytes had some precision failures (5 pesticides, 29 PCBs). Only PCB 189 and mirex had rejections due to precision failures (two batches).

Holding Times

Nineteen percent of the results (5,441 out of 37,707 total results) were classified as estimated due to holding time exceedances. These results consisted of organochlorine

pesticides, PCBs, and total mercury analyses. Tissue samples analyzed for organochlorine pesticides and PCBs exceeded either the 12 month holding time criteria between collection and extraction or the 40 day holding time criteria from extraction to analysis. Tissue samples analyzed for total mercury and selenium exceeded the 12 month holding time criteria between collection and analysis.

Assessment Thresholds

This report employed two types of thresholds for concern for pollutants in sport fish tissue that were developed by OEHHA (Klasing and Brodberg 2008): Fish Contaminant Goals (FCGs) and Advisory Tissue Levels (ATLs) (Table 4).

FCGs, as described by Klasing and Brodberg (2008), are "estimates of contaminant levels in fish that pose no significant health risk to humans consuming sport fish at a standard consumption rate of one serving per week (or eight ounces [before cooking] per week, or 32 g/day), prior to cooking, over a lifetime and can provide a starting point for OEHHA to assist other agencies that wish to develop fish tissue-based criteria with a goal toward pollution mitigation or elimination. FCGs prevent consumers from being exposed to more than the daily reference dose for non-carcinogens or to a risk level greater than 1x10⁻⁶ for carcinogens (not more than one additional cancer case in a population of 1,000,000 people consuming fish at the given consumption rate over a lifetime). FCGs are based solely on public health considerations without regard to economic considerations, technical feasibility, or the counterbalancing benefits of fish consumption." For organic pollutants, FCGs are lower than ATLs.

ATLs, as described by Klasing and Brodberg (2008), "while still conferring no significant health risk to individuals consuming sport fish in the quantities shown over a lifetime, were developed with the recognition that there are unique health benefits associated with fish consumption and that the advisory process should be expanded beyond a simple risk paradigm in order to best promote the overall health of the fish consumer. ATLs provide numbers of recommended fish servings that correspond to the range of contaminant concentrations found in fish and are used to provide consumption advice to prevent consumers from being exposed to more than the average daily reference dose for non-carcinogens or to a risk level greater than 1x10⁻⁴ for carcinogens (not more than one additional cancer case in a population of 10,000 people consuming fish at the given consumption rate over a lifetime). ATLs are designed to encourage consumption of fish that can be eaten in quantities likely to provide significant health benefits, while discouraging consumption of fish that, because of contaminant concentrations, should not be eaten or cannot be eaten in amounts recommended for improving overall health (eight ounces total, prior to cooking, per week). ATLs are but one component of a complex process of data evaluation and interpretation used by OEHHA in the assessment and communication of fish consumption risks. The nature of the contaminant data or omega-3 fatty acid concentrations in a given species in a water body, as well as risk communication needs, may alter strict application of ATLs when developing site-specific advisories. For example, OEHHA may recommend that consumers eat fish containing low levels of omega-3 fatty acids less often than the ATL table would suggest based

solely on contaminant concentrations. OEHHA uses ATLs as a framework, along with best professional judgment, to provide fish consumption guidance on an ad hoc basis that best combines the needs for health protection and ease of communication for each site."

For methylmercury and selenium, the 3 serving and 2 serving ATLs are lower than the FCGs. Consistent with the description of ATLs above, the assessments presented in this report are not intended to represent consumption advice.

The OEHHA thresholds do not take into consideration effects of contaminants on wildlife. Exposures and risks to wildlife, such as fish-eating birds, at the concentrations observed in California lakes, are likely to be higher than for humans in some instances. Due to the limits of the funding for this survey of bioaccumulation in California lakes, assessment of risks to wildlife was beyond the scope of this study. A different sampling design, focusing on different indicators (e.g., different fish species – either wildlife prey or fish that are themselves sensitive to pollutant effects – or avian eggs) would be desired to accurately evaluate exposure and risks in sensitive wildlife species. Assessment of the impact of bioaccumulation on aquatic life, though not feasible with the current level of funding, is considered a significant concern and would be evaluated if funding of this program increases sufficiently in the future.

Data Analysis

In comparing results to methylmercury thresholds, concentrations in individuals and location composites were used in a combined assessment. For individual largemouth bass, sufficient data were collected to estimate length-standardized methylmercury concentrations using analysis of covariance with a general linear mixed model. For other species, arithmetic mean concentrations of results for individuals were calculated. Geometric means were not used because the small numbers of concentrations being averaged (usually of composite samples) spanned a narrow range (Costa 2009), and because average data for individual fish were compared to equal-weight composite pooled samples.

In previous studies, largemouth bass have exhibited a strong size:methylmercury relationship when collected over a wide (spanning 150 mm or more) size range (Melwani et al. 2007; Davis et al. 2008), and have provided reasonable estimations of size-standardized methylmercury concentrations. The general linear model employed here (PROC MIXED in SAS v. 9.1; Littell et al. 1996) used a maximum likelihood approach (Burnham and Anderson 2002) to evaluate the "best" regression model from which to estimate methylmercury concentrations. Once the "best" model was selected, the relationship between fish length and methylmercury concentrations among lakes was tested to obtain the appropriate parameter estimates. The method employed dummy variables to determine differences in means, slopes, and curve shapes. The resulting regression equations were used to calculate predicted methylmercury concentrations (mean and 95% confidence interval) for each lake in a 350 mm (total length) largemouth bass. The 350 mm value was selected to represent the middle of the typical size distribution above the legal limit of 305 mm (12 in) for largemouth bass in California.

Next, average methylmercury concentrations (whether standardized for length or not) were combined with methylmercury concentrations based on composites, by taking the maximum average concentration among species. If multiple composites were analyzed for a given lake and species, the average of these data were calculated prior to taking the maximum among species. These concentrations were then compared to the thresholds selected for methylmercury (Table 4).

To compare concentrations for organic contaminants and selenium to thresholds, the concentrations in bottom species from lake-wide composites, as well as any location composites were used. Organics and selenium were not measured in individual fish. As with methylmercury, these composite results were compared with the OEHHA thresholds.

To assess statewide condition, the same approach described above was taken. Only the randomly selected lakes provide an unbiased assessment of statewide condition. These lakes were selected using the GRTS approach, and are most appropriate for performing a CDF analysis of lake condition across the state. For methylmercury, the composites and individuals from random lakes were used. For organic contaminants and selenium, the average of composites from small lakes and lake-wide or location composites from medium to large lakes were used. For all contaminants, where multiple species were sampled at a given lake, the maximum average concentration among species was selected.

Candidates for 303(d) Listing

One of the objectives of this survey was to provide information that could be used in evaluating whether a given lake should be included on the 303(d) List for each pollutant. The sampling design was developed specifically to address this objective. To meet listing requirements in a cost-effective manner, additional samples were analyzed for lakes where an initial analysis of a lakewide composite sample showed that concentrations approached a threshold.

This report does not, however, present an assessment for the purposes of 303(d) listing determinations. There are several reasons for this. First, other data and other considerations will factor in to decisions made by the Regional Boards on listing. Second, with the availability of new thresholds recently developed by OEHHA, it is unclear which thresholds will be used by the State and Regional Boards for 303(d) evaluation. Third, the State and Regional Boards will have to decide whether to modify the requirement for replicate samples to possibly include replicates collected from the same date and location.

Maps showing which lakes are candidates for 303(d) listing given different assumptions about thresholds and replication can be generated upon request. Please contact Jay Davis (jay@sfei.org) for further information.

Mapping and GIS Methods

The map figures were designed using ESRI ArcInfo 9.1 software and are in a California Teale Albers NAD 83 Projection. A connection to the GIS from the SWAMP Tissue Database 2.5 (Microsoft Access 2003) was established to display the results of queries that calculated concentrations.

RESULTS AND DISCUSSION

In the first year of this screening study, over 6000 fish from 18 species were collected from 152 lakes and reservoirs in California (Figure 1a-c, Tables 1a,b). As described in the previous section, results for PCBs and chlordanes in some samples (14 for PCBs and 4 for chlordanes) did not pass QA review due to blank contamination and are being reanalyzed. Data for these analytes in the affected lakes are not included in this report, but will be included in next year's report on the full two-year dataset. Due to these problems, smaller datasets are presently available for PCBs (138 lakes) and chlordanes (4 lakes), and for the net assessment of contamination in each lake (16 lakes – two lakes had problems with both PCBs and chlordanes).

A concise summary of the data for each lake is provided in Appendix A. More detailed summaries are provided in Appendices B (average and composite concentrations for all samples) and C (results for methylmercury analyses on individual fish). Excel files containing these tables are available from SFEI (contact Jay Davis, jay@sfei.org). The complete dataset is available from the SWAMP data management team at Moss Landing Marine Laboratories. The complete dataset includes data on QA analyses, additional ancillary information, and data for blind duplicates that may be of use in 303(d) determinations. All data collected for this study are maintained in the SWAMP database which is managed by the data management team at Moss Landing Marine Laboratories. The SWAMP database also stores water quality, tissue, and bioassessment data along with the associated quality assurance samples. Tissue data will soon be available on the web at http://www.ceden.org/. Until then contact Cassandra Lamerdin (clamerdin@mlml.calstate.edu) for more information on the complete data set.

Net Assessment of Lake Contamination

"Net assessment" refers to the overall degree of contamination of each lake with consideration of all measured pollutants for which thresholds are available (methylmercury, PCBs, dieldrin, DDTs, chlordanes, and selenium). Analytical results for all pollutants at each lake were compared to their respective thresholds of concern. The thresholds selected for these comparisons were OEHHA's (Klasing and Brodberg, 2008) fish contaminant goals and advisory tissue levels (ATLs) (Table 4). The lowest available threshold was used for each pollutant. The intent of this assessment is to answer the following question (one aspect of Management Question 1): Which of the sampled lakes appear to be below all thresholds of concern based on data obtained from this study? Lakes with all samples below thresholds are considered to have tested "clean" in this screening survey.

Only 21 of the 136 lakes (15%) with complete data from 2007 had all samples below all thresholds for all pollutants (Figure 2). Methylmercury was the pollutant primarily responsible for so many lakes having at least one sample above thresholds. Overall, 74% of the 152 lakes sampled had a methylmercury concentration above the lowest threshold for methylmercury (the 0.07 ppm three serving ATL). In the random sample of 50 lakes, 80% of the lakes had a species with an average methylmercury concentration higher than 0.07 ppm (Figures 3a,b). The 95% confidence interval for this estimate was 68 – 91%. For the random sample, the degree of impact could also be expressed on an areal basis, but the percentage was similar (78%). For targeted lakes (n=102), 70% had a species average higher than 0.07 ppm (Figure 3b). Most (61%) of the northern California trout lakes were below 0.07 ppm, and only 3% were above 0.44 ppm (Table 6). This was in sharp contrast to lower elevation lakes in northern California, which had only 4% below 0.07 ppm and half of the lakes (50%) above 0.44 ppm. Concentrations in Southern California were intermediate, with 31% below 0.07 ppm and 15% above 0.44 ppm.

PCBs had a secondary role in causing lakes to exceed thresholds. The lowest threshold for PCBs was the FCG (3.6 ppb). For PCBs, 37% of the 138 lakes with results reported for year 1 were above this threshold: 20% of the random lakes and 43% of the targeted lakes (Figures 4a,b). Southern California had a higher percentage of lakes with at least one sample above 3.6 ppb (60%) than lower elevation lakes in northern California (41%) and northern California trout lakes (7%) (Table 7).

Other pollutants caused lower percentages of samples to exceed thresholds:

- dieldrin exceeded the 0.46 ppb FCG in at least one sample in 21% of 152 lakes;
- DDTs exceeded the 21 ppb FCG in at least one sample in 17% of 152 lakes;
- chlordanes exceeded the 5.6 ppb FCG in at least one sample in 10% of the 148 lakes with data; and
- selenium exceeded the 2500 ppb three serving ATL in at least one sample in 2% of the 120 lakes with data.

All of these pollutants were below thresholds in all northern California trout lakes and had similar percentages of samples above FCGs in southern California and lower elevation lakes in northern California (Tables 8 – 11).

With methylmercury being the pollutant primarily exceeding the ATL, factors affecting methylmercury concentrations were important in determining the overall pattern of lake contamination. One of the characteristics that most of the apparently clean lakes had in common was the absence of largemouth bass. Largemouth bass is a high trophic level species that usually accumulates high concentrations of methylmercury relative to other species. Only one of the 21 clean lakes had a largemouth bass sample (#30 Lake of the Pines in Region 5). This lake stands out as having exceptionally low methylmercury contamination.

Most of the clean lakes were in regions at higher elevations (particularly in the Sierra Nevada), beyond the range where largemouth bass and other warm water species

(common carp and channel catfish) are abundant, and where trout species predominate (rainbow trout, brown trout, and Eagle Lake trout were collected in this survey). Trout were sampled at 15 of the 21 clean lakes (14 had rainbow trout and one had brown trout). Trout generally occupy a lower trophic position and accumulate lower concentrations of methylmercury and other pollutants, though exceptions to this pattern occur and were observed in this study (discussed further below). Another factor that probably contributes to lower observed concentrations in trout is that, in many lakes, recently planted hatchery fish are part of the catch. A previous study found that hatchery trout consistently had very low concentrations of methylmercury (rainbow trout from four hatcheries all had less than 0.023 ppm – Grenier et al. 2007). It is important to note that resident, self-sustaining trout populations in these lakes are likely to have higher concentrations than the hatchery fish that are most readily collected. The potential influence of hatchery trout on the results is discussed further in the Methylmercury section below.

Another group of clean lakes was in warmer waters at low elevations where largemouth bass commonly occur, but where bass were not collected. The species sampled at these lakes (common carp, channel catfish, black crappie [Pomoxis nigromaculatus], and bluegill [Lepomis macrochirus]) tend to occupy a lower trophic position than largemouth and accumulate lower concentrations of methylmercury. The two apparently clean lakes in southern California and the one clean lake in Region 2 fell into this category.

Lakes that were classified in the "clean" category based on this one survey are not necessarily entirely free of bioaccumulation problems. Most of these apparently clean lakes did not yield the species that tend to have high pollutant concentrations. Whether the lakes that tested clean in this survey can really be considered entirely clean or not depends on whether high methylmercury species such as largemouth bass or self-sustaining trout populations are really absent from these lakes. While the methods used to collect fish in this survey are generally effective for largemouth bass and other black bass species, it is possible that such species were present in some lakes where they were not collected, especially in the low elevation lakes where other warm water species were collected. Nevertheless, falling into the green category in this survey is a positive outcome, indicating that the most readily caught species in a lake have pollutant concentrations that are below thresholds for concern.

Lakes that had one or more locations above a threshold (red symbols in Figure 2) are candidates for additional monitoring and perhaps advisory development. Further prioritization of these lakes is discussed below.

Methylmercury

Comparison to Thresholds

Methylmercury is the pollutant that poses the most widespread potential health risks to consumers of fish caught from California lakes. As discussed in the previous

section, methylmercury concentrations measured in this study were very frequently higher than the lowest OEHHA threshold for methylmercury – 0.07 ppm – a concentration at which OEHHA would consider recommending consumption of less than three servings per week. Furthermore, methylmercury was the only pollutant that frequently reached concentrations high enough that OEHHA would consider recommending <u>no</u> consumption of the contaminated species (0.44 ppm). Overall, 39 of the 152 lakes surveyed (26%) had a species with an average concentration exceeding 0.44 ppm. For the random lakes, 23% were above 0.44 ppm (18% on an areal basis) (Figure 3a), while 26% of the targeted lakes were above this threshold (Figure 3b).

One important finding from year 1 is that very few California lakes contain predatory fish, such as largemouth bass, with low concentrations of methylmercury (Figure 5). The average (size-adjusted) concentrations observed in the lakes that were below thresholds were 0.07 ppm in Lake of the Pines (Region 5), 0.03 ppm in Lake Calabassas (Region 4), 0.01 ppm in Toluca Lake (Region 4), and 0.07 ppm in Prado Lake (Region 8). These low concentrations may be due to variation in ecosystem factors such as water chemistry, productivity, trophic dynamics, wetland presence, or others; or due to variation in sources, such as the absence of mining influence. The low concentrations observed at these lakes indicate that it is indeed possible for lakes in the California landscape to not have excessive bioaccumulation of methylmercury, and that a management goal for at least some lakes may be to attain concentrations of this magnitude.

Spatial Patterns

Methylmercury concentrations across the state varied at a regional scale (Figure 6). In northern California, low concentrations were commonly observed in high elevation lakes in the Sierra Nevada and Trinity Alps. The highest species averages observed in most of these lakes were below the three-serving ATL (0.07 ppm). Trout (mostly rainbow trout, but a few lakes had brown trout or Eagle Lake trout) were the most commonly caught species in these lakes, and, as discussed above, tend to accumulate lower methylmercury concentrations than largemouth bass. For the 31 northern California trout lakes sampled, 61% had a maximum species average below 0.07 ppm, another 26% were between 0.07 and 0.15 ppm, and only one of these lakes (3%) had a species average above 0.44 ppm – Hetch Hetchy Reservoir (Table 12).

The results from Hetch Hetchy Reservoir illustrate an important point about trout lakes – the concentrations measured in this screening survey may be heavily influenced by recently planted hatchery fish and may not be representative of self-sustaining populations of fish that may also be present in these lakes. Hetch Hetchy Reservoir was anomalous among the trout lakes with methylmercury concentrations of 0.96 and 0.54 ppm in composites of brown trout from two distinct locations (Figure 7). One other lake (Loon Lake) also had relatively high concentrations in two composites of brown trout (0.50 and 0.30 ppm). Brown trout from the other six lakes where they were collected had low concentrations (all around 0.10 ppm or less). While the high concentrations in Hetch

Hetchy indicate that the food web in this reservoir is relatively contaminated with methylmercury, two other factors also probably contribute to the anomalous results.

First, the brown trout population in Hetch Hetchy is self-sustaining. Hetch Hetchy has not been stocked in many years (Jay Rowan, California Department of Fish and Game, personal communication). As mentioned above, many trout lakes are stocked with fish from hatcheries that past work (Grenier et al. 2007) has indicated are probably low in methylmercury. Hetch Hetchy may be anomalous because the brown trout collected were lifelong residents that had more time to accumulate methylmercury concentrations that are representative of the Hetchy Hetchy food web. Boles (2007) also observed relatively high methylmercury concentrations (0.35 ppm in a composite of five fish) in brown trout from another reservoir (Sly Creek Reservoir in Butte County) with a self-sustaining population. These findings suggest that although the results obtained in this screening study do probably accurately portray concentrations in the predominant catch taken by anglers, they may not be accurate indicators of the degree of contamination of the food webs or self-sustaining fish populations in lakes where extensive planting of hatchery fish occurs.

A second factor that could contribute to the high concentrations in brown trout from Hetchy Hetchy Reservoir and Loon Lake is that brown trout are known to switch to piscivory as they get older (Moyle 2002). The brown trout samples with high methylmercury were all above 400 mm in average length, while the samples with lower methylmercury were all below 400 mm (Figure 7).

In contrast to the northern California trout lakes, methylmercury concentrations in lower elevation (below 2000 ft) lakes in northern California (Table 6, Figure 6) were almost always higher than the three-serving per week ATL (0.07 ppm), and frequently higher than the no consumption ATL (0.44 ppm). Of the 56 lower elevation lakes sampled in northern California, 50% had a maximum species average above 0.44 ppm, another 34% were between 0.22 and 0.44 ppm, and only two (4%) lakes in this region had a species average below 0.07 ppm. The three lakes that had a methylmercury concentration at or below 0.07 ppm were Lago Los Osos in Region 2 and Lake of the Pines in Region 5. Largemouth bass were not caught at Lago Los Osos – only channel catfish were collected. Lake of the Pines was the only lake in northern California where largemouth bass were collected that had an average concentration at a standard size of 350 mm of 0.07 ppm or lower. Interestingly, the concentration measured at this lake was in sharp contrast to concentrations in 350 mm largemouth at two adjacent lakes: Lake Combie immediately to the south at 0.78 ppm and Zayak/Swan Lake to the north at 0.98 ppm.

Although methylmercury concentrations were generally not as high in southern California, the methylmercury problem is not confined to northern California and its well-known mining regions. Most of the 55 lakes in southern California were between 0.07 and 0.44 ppm (55%), but 15% had a maximum species average above 0.44 ppm (Table 6). Average concentrations as high as 0.95 ppm were observed (Crystal Lake). The remaining lakes (31%) in this region had a species average below 0.07 ppm (Table 6,

Figure 6). Largemouth bass were collected at only three of the 17 lakes that were below 0.07 ppm in southern California: Lake Calabassas, Toluca Lake, and Prado Lake.

Priorities for Further Assessment

Lakes with average methylmercury concentrations of one or more species above 0.44 ppm should be considered high priorities for further assessment to determine the need for consumption guidelines and management actions. Many lakes had concentrations well above the 0.44 ppm threshold (Table 12). Lake Pillsbury had the highest species average concentration in the state (1.31 ppm in 350 mm largemouth bass), and the highest concentration for an individual fish – 4.08 ppm in a very large (559 mm) largemouth bass. Other lakes with a species average concentrations above 1 ppm included (all are in 350 mm largemouth bass unless otherwise noted): Cosumnes River in Region 5 (1.15 ppm); Chesbro Reservoir in Region 3 (1.04 ppm); Lake Nacimiento in Region 3 (1.00 ppm in smallmouth bass [not size-adjusted]); and Upper San Leandro Reservoir in Region 2 (1.01 ppm). Table 12 shows the data for samples at the 37 lakes that had a species average above 0.44 ppm based on either composite samples or the ANCOVA results. Consumption guidelines have already been issued for 10 (27%) of these lakes, but 27 (73%) do not have guidelines.

Implications Regarding Sources

Although evaluating sources is not a primary goal of the study, the results of this two-year survey of methylmercury and other pollutants in sport fish may yield valuable information on sources of the contamination and other factors that influence bioaccumulation. At least a preliminary analysis of this topic may be illuminating and will be performed in the final report covering both years of the study. The analysis in the final report will attempt to explain some of the interesting patterns observed in year 1.

The extensive statewide dataset generated in this study may shed some light on the relative importance of sources of mercury such as historic mining activity and atmospheric deposition. The low methylmercury concentrations observed at some lakes indicate that atmospheric deposition at a broad geographic scale is not large enough to cause excessive bioaccumulation in all California lakes. On the other hand, the broad distribution of the methylmercury problem throughout California suggests that atmospheric deposition may play a major role. Regarding the influence of mining, the greater prevalence of high concentrations in northern California appears to be consistent with the larger amount of mercury and gold mining activity in that region (Figure 8). It should be noted, however, as indicated on Figure 8, that gold and silver mining were also extensive in southern California, with a relatively dense cluster of historic mine sites in the area of Region 4 with most of the southern California lakes above 0.44 ppm. A finer scale analysis of lake characteristics, upstream mining activity, other sources (e.g., landfills, wastewater discharges, incinerators, gas pipelines, electrical equipment, and pesticides), and other factors will be needed to attempt to resolve these questions.

PCBs

Comparison to Thresholds

PCBs (measured as the sum of 55 congeners) were second to methylmercury in reaching concentrations posing potential health risks to consumers of fish caught from California lakes. However, far fewer lakes had PCB concentrations exceeding OEHHA's higher risk thresholds (Table 7). Overall, only two of the 138 lakes assessed in year 1 (1.47%) had a species with an average concentration high enough that OEHHA would consider recommending no consumption of the contaminated species (120 ppb). The majority of these lakes (87%) were below the three serving ATL for PCBs (21 ppb). However, 37% exceeded the lowest OEHHA threshold – the FCG of 3.6 ppb.

The frequency distributions were different for random and targeted lakes. This was due to the relatively extensive sampling of Region 4, the region with the highest PCB concentrations. For the random sampling, 20% of the sampled lakes were above 3.6 ppb, while 43% were above this threshold for the targeted lakes (Figures 4a,b). For the random lakes, the percentages expressed on an areal basis were very similar to those expressed on a per lake basis.

Spatial Patterns

PCB concentrations across the state varied at a regional scale (Table 7, Figure 9). As for methylmercury, in northern California, low concentrations were commonly observed in high elevation lakes in the Sierra Nevada and Trinity Alps. The vast majority of species averages observed in these lakes were below the FCG (3.6 ppb). For the 30 northern California lakes where trout were collected, 93% had a maximum species average below 3.6 ppb, one lake (3%) was between 3.6 and 21 ppb (the 3 serving ATL), one lake (3%) was between 21 and 42 ppb (the 2 serving ATL), and none were above 42 ppb. The highest species average measured in this region was 28 ppb in a brown trout sample from Silver Lake in Region 6.

PCB concentrations were greater than the trout lakes in low elevation (below 2000 ft) lakes in northern California (Table 7, Figure 9). Of the 54 low elevation lakes sampled in northern California, 59% had a maximum species average below 3.6 ppb, 28% were between 3.6 and 21 ppb, 4% were between 21 and 42 ppb, 9% were between 42 and 120 ppb, and none were above 120 ppb. Average concentrations at two low elevation lakes from northern California were among the highest concentrations measured in the state (Table 13): Lake Chabot in San Leandro in Region 2 (98 ppb) and San Luis Reservoir in Region 5 (85 ppb).

Southern California was the region with the highest PCB concentrations. Of the 45 lakes in southern California with data reported, 40% had a maximum species average below 3.6 ppb, 36% were between 3.6 and 21 ppb, 9% were between 21 and 42 ppb, 11% were between 42 and 120 ppb, and two lakes (4%) were above 120 ppb (Table 7). Average concentrations at four lakes from southern California were among the highest concentrations measured in the state (Table 13): Pyramid Lake (238 ppb in brown

bullhead), Elderberry Forebay (131 ppb in channel catfish), and Echo Lake (101 ppb in common carp) in Region 4; and Silverwood Lake (93 ppb in largemouth bass). Pyramid Lake and Elderberry Forebay were the two lakes in the state exceeding the 120 ppb no consumption ATL. The PCB concentrations observed in largemouth bass in Silverwood Lake are exceptionally high for this species, and much higher than those measured largemouth bass from Pyramid Lake where the higher lipid, bottom-feeding species (brown bullhead) reached the maximum concentrations observed in the entire dataset.

Priorities for Further Assessment

Using the same criterion that was employed for methylmercury (i.e., exceedance of the no consumption ATL - 120 ppb for PCBs) only two lakes (in contrast to 37 for methylmercury) stand out as high priorities for further assessment to determine the need for consumption guidelines and management actions. Pyramid Lake in Region 4 had the highest species average by far for PCBs in the state (238 ppb), and the highest concentration in a sample (416 ppb in a composite sample). Elderberry Forebay, a lake just 10 miles away from Pyramid Lake, was the other lake with an average concentration exceeding 120 ppb (131 ppb). The high concentrations in largemouth bass at Silverwood Lake suggest that this water body may also warrant further investigation. Echo Lake and Peck Road Water Conservation Park also had relatively high concentrations in largemouth bass (48 ppb and 39 ppb, respectively). Consumption guidelines have not been issued for these lakes.

Implications Regarding Sources

The geographic distribution of PCBs measured in California sport fish provides an indication of the location and nature of the principal sources of these chemicals. A review of historic bioaccumulation monitoring of PCBs in California (Davis et al. 2007) found that high concentrations of PCBs tended to occur in areas of historic use or maintenance of electrical equipment. These areas tend to be concentrated in urban centers with high amounts of industrial activity, but also occur in scattered areas across the landscape where electrical equipment or other PCB-containing equipment was used. The many hydroelectric facilities in the state are potential sites of past or present PCB contamination. Similar to methylmercury, significant variation exists among species in their tendency to accumulate PCBs, with high-lipid bottom-feeders like common carp, channel catfish, and brown bullhead accumulating the highest concentrations. Because of this interspecific variation, a map of concentrations in common carp and channel catfish provides a clearer picture of spatial variation (Figure 10). The patchy distribution of PCBs across the state, with lakes with low concentrations observed in most areas and scattered lakes with much higher concentrations, is consistent with contamination by local sources. One possible exception is in the Los Angeles region, where the very high prevalence of lakes above the FCG may suggest an elevated signal of regional atmospheric deposition. Other urban sources, such as urban runoff and landfill leachates may also contribute to this regional pattern.

Other Pollutants With Thresholds

OEHHA (Klasing and Brodberg 2008) developed thresholds for four other pollutants that were analyzed in this survey: dieldrin, DDT, chlordane, and selenium. Concentrations of these pollutants infrequently exceeded any threshold, and never exceeded the no consumption ATLs. The high elevation trout lakes of northern California never exceeded any threshold for these pollutants. Results for these pollutants are briefly summarized below.

Dieldrin

The maximum species averages for dieldrin were below the lowest threshold (the 0.46 ppb FCG) in 79% of all the lakes sampled, including 100% of the northern California trout lakes, 70% of the northern California low elevation lakes, and 75% of the southern California lakes (Figure 11, Table 8). None of the ATL thresholds were exceeded in any part of the state. The highest species average measured was 6.6 ppb in common carp from San Luis Reservoir. The highest concentration measured in any sample was 11.3 ppb in a common carp composite from San Luis Reservoir. Relative to methylmercury and PCBs, none of the lakes sampled appear to be a high priority for further assessment or action based on dieldrin concentrations.

DDTs

The maximum species averages for DDTs were below the lowest threshold (the 21 ppb FCG) in 83% of all the lakes sampled, including 100% of the northern California trout lakes, 75% of the northern California lower elevation lakes, and 78% of the southern California lakes (Figure 12, Table 9). Only one lake exceeded the 3 serving ATL threshold for DDTs (520 ppb): Pinto Lake in Region 3, which had a concentration of 557 ppb in a common carp composite. Relative to methylmercury and PCBs, none of the lakes sampled appear to be a high priority for further assessment of human health risks due to DDT contamination. Risks to wildlife from DDT contamination in some lakes, however, are likely to be significant. Based on the degree of contamination observed in this survey, DDT would be expected to exceed thresholds for effects on raptor reproduction in some lakes.

Chlordanes

The maximum species averages for chlordanes were below the lowest threshold (the 5.6 ppb FCG) in 90% of all the lakes sampled, including 100% of the northern California trout lakes, 86% of the northern California lower elevation lakes, and 84% of the southern California lakes (Figure 13, Table 10). None of the ATL thresholds were exceeded in any part of the state. The highest species average measured was 60 ppb in common carp from Lake Lindero in Region 4. The highest concentration measured in any sample was 87 ppb in a common carp composite from Lake Lindero. Relative to methylmercury and PCBs, none of the lakes sampled appear to be a high priority for further assessment or action based on chlordane concentrations.

Selenium

The maximum species averages for selenium were below the lowest selenium threshold (the 3 serving ATL of 2500 ppb) in 98% of all lakes sampled, including 100% of the northern California trout lakes, 100% of the northern California lower elevation lakes, and 95% of the southern California lakes (Figure 14, Table 11). Only Ramer Lake (3020 ppb) and Salton Sea (2580 ppb) in Region 7 and Lake Lindero (2790 ppb) in Region 4 exceeded the 2500 ppb threshold. The highest species average measured was 3020 ppb in common carp from Ramer Lake in Region 7. The highest concentration measured in any sample was 3850 ppb in a common carp composite from Ramer Lake. Relative to methylmercury and PCBs, none of the lakes sampled appear to be a high priority for further assessment or action based on chlordane concentrations.

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